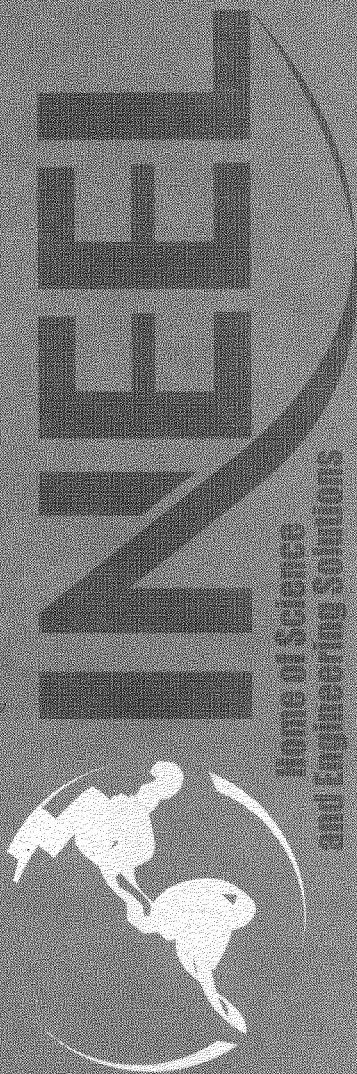


Treatment Technologies for the OU 7-10 Stage III Project

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August 2003*



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August 2003

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ABSTRACT

Mixed waste treatment technologies have been investigated as alternatives to incineration to determine their feasibility when used on retrieved waste from Operable Unit 7-10 (Pit 9). It was necessary for technologies to be sufficiently mature to allow implementation on the excavated waste within a short period of time. Treatability study testing is anticipated for each of the technologies for each excavated waste stream. However, technologies that require extensive development and proof-of-principle testing have not been considered. Three different steam-reforming technologies have been investigated and were identified in *Evaluation of Alternative Nonflame Technologies for Destruction of Hazardous Organic Waste*. The same evaluation process has been performed on thermal desorption solvated electron technologies that were not previously discussed or evaluated in this report. Technologies were screened for effectiveness, implementability, and cost in accordance with Comprehensive Environmental Response, Compensation, and Liability Act guidance.

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ACRONYMS

COC	contaminant of concern
HEPA	high-efficiency particulate air
HSE	heated screw evaporator
INEEL	Idaho National Engineering and Environmental Laboratory
LDR	land disposal restriction
OU	operable unit
PCB	polychlorinated biphenyl
RCRA	Resource Conservation and Recovery Act
RFP	Rocky Flats Plant
SET	Solvated Electron Technology
THOR	Thermal Organic Reduction
TRU	transuranic
VOC	volatile organic compound
WAC	waste acceptance criteria

Treatment Technologies for the OU 7-10 Stage III Project

1. INTRODUCTION

This report is an extension of *Evaluation of Alternative Nonflame Technologies for Destruction of Hazardous Organic Waste* (INEEL 1997). It provides an update on promising technologies that could be implemented at Operable Unit (OU) 7-10 of the Subsurface Disposal Area in the Radioactive Waste Management Complex to support the OU 7-10 Stage III design. In addition, technologies commercially available but not considered in the original report are identified.

This document identifies technologies sufficiently mature (beyond bench scale) for treating (destroying, reducing, or stabilizing) Resource Conservation and Recovery Act (RCRA) components of the waste. Transuranic (TRU) waste with activities greater than 100 nCi/g that meets waste acceptance criteria (WAC) will be shipped to the Waste Isolation Pilot Plant near Carlsbad, New Mexico. For waste streams that have activity levels equal to and less than 100 nCi/g, waste eventually will be returned to OU 7-10. Mixed waste streams that have activity levels equal to and less than 100 nCi/g will be treated for the organic contaminants of concern (COCs) so that remaining organic contamination levels meet delisting levels identified in the Record of Decision. The inorganic contaminants, nitrates and cyanides will be decharacterized or destroyed before returning waste to the pit. Any RCRA metals that exceed the toxicity characteristic levels will be stabilized after organic contaminant removal and destruction so that the RCRA inorganic material does not exceed leaching criteria identified in the land disposal restrictions (LDRs). The treated material then will be returned to OU 7-10 for disposal.

Technologies not identified in the earlier evaluation (INEEL 1997) have been presented in the same format given in Appendix A of that document. Technologies identified in the earlier document are updated to show their present status. The ranking process for these technologies has not been performed.

The earlier report defines the distinction between thermal destruction and thermal extraction technologies. Based on the definition given, it is concluded that thermal desorption and other low temperature separation processes (steam reforming) are viable technologies that should be further investigated for treatment of the OU 7-10 waste streams. Nonthermal technologies also are identified in the 1997 report. The list of technologies potentially capable of treating OU 7-10 waste and readily available for implementation is extended in this document.

2. OPERABLE UNIT 7-10 WASTE

2.1 Pit Operations

OU 7-10 was operated as a waste disposal pit from November 8, 1967, to June 9, 1969. Approximately 7,080 m³ (250,000 ft³) of overburden, 4,247 m³ (150,000 ft³) of packaged waste, and 9,911 m³ (350,000 ft³) of soil between and below the buried waste were in OU 7-10 at the time of closure (DOE-ID 1993). The pit was excavated to basalt bedrock between 4 and 6.4 m (13 and 21 ft) deep (Ebasco 1990). Soil placed over the bedrock provided a level surface for placing the waste. The layer varies in depth because the bedrock is uneven but may be as much as several feet thick. Soil placed over the waste was intended to provide a 0.9-m (3-ft) overburden layer. Because of maintenance for subsidence from various floods, the overburden layer is considered to vary from 0.8 m (2.5 ft) to more than 1.5 m (5 ft) depending on the location.

The waste in OU 7-10 is primarily TRU waste generated at the Rocky Flats Plant (RFP) totaling 3,115 m³ (110,000 ft³) with additional low-level waste from Idaho National Engineering and Environmental Laboratory (INEEL) generators totaling 1,133 m³ (40,000 ft³). At the time of disposal of the RFP waste, TRU-contaminated waste was defined as having a TRU activity level greater than 10 nCi/g compared to the present day level of 100 nCi/g. Five plutonium radionuclides (Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242) and the isotope Am-241 compose 99.9% of the radiological activity originally disposed of in OU 7-10. Operable Unit 7-10 also contains U-234, U-235, and U-238 primarily in the form of depleted uranium. Other categories of radionuclides in OU 7-10 are small quantities of mixed activation products and mixed fission products from INEEL generators.

2.2 Rocky Flats Waste

Waste from RFP consisted primarily of drums of Series 74 sludge, drums and wooden boxes of assorted waste, and cardboard cartons containing empty contaminated drums. Shipping records indicate that 2,077 55-gal drums of Series 74 sludge was buried in OU 7-10. Five types of sludge drum waste were shipped to the INEEL from RFP as Series 74 sludge drums: 741, first stage sludge; 742, second stage sludge; 743, organic sludge; 744, solidified liquids; and 745, evaporator salts (nitrates). Series 741 and 742 are uncemented sludge, and Series 744 are cemented sludge. All Series 74 sludge was placed inside double polyethylene bags within a 55-gal drum.

Series 741 and 742 sludge consist of water and a precipitate of hydrated oxides of iron, magnesium, aluminum, silicon, plutonium, americium, and depleted uranium. Each drum of 741 and 742 sludge was layered with 18.2–22.7 kg (40–50 lb) of Portland cement to absorb any free liquid. (Cement was not mixed with the sludge.) The drums of 742 sludge received from RFP before OU 7-10 closure may contain other waste items such as electric motors, polyethylene or glass containers of liquid chemical waste, mercury and lithium batteries, and small amounts of mercury in pint bottles.

Series 743 sludge consisted of a mixture of about 140 L (37 gal) of organic liquid and 45 kg (100 lb) of calcium silicate along with 4.5–9 kg (10–20 lb) of oil adsorbent. The organic liquid consists of about 47% lathe coolant ([43.5% Texas Regal oil, 56.5% carbon tetrachloride], 10% degreasing agents [1,1,1-trichloroethane], and 43% miscellaneous organic compounds). In addition, oil contaminated with polychlorinated biphenyls (PCBs) may have been processed with other organic waste in the Series 743 sludge.

In each drum containing Series 744 sludge, approximately 98 L (26 gal) of liquid waste were mixed with 86 kg (190 lb) of Portland cement and 23 kg (50 lb) of magnesia cement. Approximately 4–7

kg (10–15 lb) of additional Portland cement were placed on top of the cement mixture before it was sealed in a plastic bag. Cement was added to absorb the liquid waste into a solid matrix. The waste form contains complexing chemicals that consist of Versenes (trademark for a series of chelating agents based on ethylenediamine tetraacetic acid), alcohols, and organic acids. All liquid waste was made basic before adding them to the cement mixture.

The content of Series 745 sludge is 60% sodium nitrate, 30% potassium nitrate, and 10% miscellaneous. These miscellaneous constituents consist of sodium and potassium chlorides, dichromates, phosphates, and sulfates. Limited amounts of other waste, such as surgeons' gloves, paper, rags, and metal, may be found in these drums.

2.3 Idaho National Engineering and Environmental Laboratory-Generated Waste

Assorted waste was trucked from seven INEEL generators and buried in OU 7-10. The low-level waste was brought in various containers (e.g., dumpster, boxes, pallets, and large components) and disposed of. All waste disposals were within the contact-handled requirement (gamma activity less than 200 mrem/hour) according to shipping records, except for items in Table 1 of Einerson and Thomas (1999). However, waste containers may no longer be intact. This could require remote handling for some buried waste.

Further details of the waste streams buried in OU 7-10 can be found in Einerson and Thomas (1999), Josten and Thomas (2000).^{a, b, c}

a. R. W., Thomas, INEEL, Interdepartmental Communication to J. C. Okeson, INEEL, "Graphite, Filter, and Potentially Pyrophoric Wastes in Pit 9 for OU 7-10 Extended Probe Hole Activities—RWT-02-00," June 8, 2000.

b. R. W., Thomas, INEEL, Interdepartmental Communication to D. E. Wilkms, INEEL, "Pit 9 Inventory Data Supporting OU 7-10 Stage I Air Emissions Evaluation—RWT-07-99," October 14, 1999.

c. R. W., Thomas, INEEL, Interdepartmental Communication to D. E. Wilkms, INEEL, "The Issue of Irradiated Fuel in Pit 9—RWT-06-99," September 14, 1999.

3. ASSUMPTIONS

To be able to identify and evaluate technologies for treating the OU 7-10 buried waste, the following operational assumptions enable comparisons:

1. All waste, where practical, shall be excavated from OU 7-10. Excavated waste either will be placed in 55-gal drums, standard waste boxes, or stockpiled on RCRA-approved pads before disposition. Excepted large objects may be left in the pit (see assumption 6 below).
2. The volume of waste that requires excavation shall include all packaged waste, 0.3 m (1.0 ft) of overburden soil, all interstitial soil, and all of the soil below the waste—a total of 15,447m³ (545,500 ft³) of waste and soil.
3. All waste containing TRU contaminants shall be separated into two groups: TRU waste (i.e., greater than 100 nCi/g) and that equal to or below 100 nCi/g.
4. All waste containing TRU contaminants greater than 100 nCi/g will be treated as required to meet the WAC, packaged, and sent to the Waste Isolation Pilot Plant for disposal.
5. All non-TRU waste (less than or equal to 100 nCi/g-TRU contaminants and the INEEL waste) that is remote handled (greater than 200 mrem/hour at the digface or container surface, whichever is reached first) will be grouted in place.
6. Excepted large objects either shall be removed and placed on the RCRA-approved pad to await final disposition or shall be grouted and abandoned in place.
7. Waste that contains less than 100 nCi/g-TRU contaminants and does not contain concentrations of hazardous COCs above Table 1 values shall be returned to OU 7-10 for disposal.
8. Waste that contains less than or equal to 100 nCi/g-TRU contaminants and contains any hazardous contaminant-of-concern concentration above Table 1 values shall be treated.
9. Treated waste that contains less than or equal to 100 nCi/g-TRU contaminants shall meet, before returning to the pit, the criteria that waste will:
 - a. No longer exhibit the characteristics of hazardous waste as defined in 40 CFR 261.3 (2003)
 - b. Meet Table 2 RCRA delisting levels for identified COCs (volatile organic compounds [VOCs], nitrates, and cyanides)
 - c. Be stabilized to meet toxicity characteristic leaching procedure requirements for RCRA toxic metals.

Remediation levels that must be achieved by any treatment technology on each RCRA constituent found in OU 7-10 are given in Table 2. These remediation levels are to be presented to the Idaho Department of Environmental Quality for the contaminant to be returned to the pit for disposal.

Table 1. Trimer levels for contaminants of concern

Contaminant of Concern	Treatment Trimer Level
Carbon tetrachloride (FOO 1 & F002)	0.2 mg/kg
Tetrachloroethylene (FOO 1 & F002)	1 mg/kg
Trichloroethylene (FOO 1 & F002)	0.05 mg/kg
1,1,1-trichloroethane (F001 & F002)	1,200 mg/kg
Methylene chloride	9 mg/kg
Nitrates	10,000 µg/L

Table 2. Return to pit criteria.

Contaminant of Concern	Return to Pit Criteria
Carbon tetrachloride (FOO 1 & F002)	0.2 mg/kg
Tetrachloroethylene (FOO 1 & F002)	1 mg/kg
Trichloroethylene (FOO 1 & F002)	0.05 mg/kg
1,1,1-trichloroethane (FOO 1 & F002)	1,200 mg/kg
Methylene chloride	9 mg/kg
Nitrates	10,000 µg/L
Sodium Cyanide	122 mg/kg
Potassium Cyanide	119 mg/kg
Toxicity characteristic leaching procedure metals	Stabilized to meet land disposal restrictions
Reactivity characteristic	No longer exhibits
Corrosivity characteristic	No longer exhibits
Ignitability	No longer exhibits

The chlorinated organic compounds have been identified in Einerson and Thomas (1999) as well as sodium and potassium nitrates. However, sodium and potassium cyanide may or may not be present. At least two 25-lb packs of sodium or potassium cyanide were distributed in Series 742 waste drums. Because there is no information on when these drums were shipped or if they were even disposed of in OU 7-10, no estimate of the total quantity in OU 7-10 can be attempted.

3.1 Volume Reduction

The Record of Decision for OU 7-10 (DOE-ID 1993) assumes that physical separation (soil washing) followed by chemical extraction will reduce the volume of contaminated soils. Soil washing operates on the principle that many contaminants concentrate on the surface of smaller particles because a given volume of these particles provides a greater surface area for adsorption than an equal volume of larger particles. For soil washing to be effective in reducing large volumes of contaminated soil, it is expected that the percentage of silt and clay in the soil will be approximately 10%. OU 7-10 contains approximately 80–90 % silt and clay.^d Thus, volume reductions of the expected magnitude are not achievable using soil washing alone.

d. INEEL, 2000, "Evaluation of Subsurface Disposal Area Buried Waste Cleanup Technologies (Draft)," INEEL/EXT-2000-00984, Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho, July 2000.

4. THERMAL TREATMENT TECHNOLOGIES

Two thermal treatments, steam reforming and thermal desorption, are candidate technologies for OU 7-10 waste streams because of their ability to meet regulatory requirements. Steam reforming was considered in the earlier evaluation (INEEL 1997), and recent developments are highlighted in this report. Thermal desorption was not considered in that report but is investigated here because of its applicability to the OU 7-10 waste streams.

4.1 Steam Reforming

Steam reforming obtained a high rating in the earlier document because of its effectiveness and advanced stage of development. Since the publication of that document, steam reforming has advanced even further. Steam reforming has been undergoing testing for treating sodium-bearing waste at the New Waste Calcining Facility at the INEEL. Two steam-reforming technology processes have been identified for treating the sodium-bearing waste: ThermoChem, Inc. and Studsvik.^e The Studsvik Thermal Organic Reduction (THOR) process uses fluidized bed technology to process various solid and liquid waste streams. The licensee for the Studsvik THOR process is THOR Treatment Technologies. Thermal Organic Reduction Treatment Technologies has developed a two-stage pyrolysis and steam-reforming process that can handle heterogeneous drummed waste beyond the requirements of the sodium-bearing waste. Duratek Services, Inc., (Duratek) has further developed the Synthetica Technologies steam-reforming technology to a system suitable for heterogeneous waste forms found in OU 7-10. In the steam-reforming reaction above 316°C, steam reacts with hydrocarbons in the feed to produce CO, CO₂, H₂O, H₂, and small amounts of CH₄. This combination is referred to as a synthesis gas.

4.1.1 Technology Description

The ThermoChem steam reformer (see Figure 1) consists of an indirectly heated fluidized bed in a refractory-lined reactor vessel. The fluidizing medium is steam for mixing and heat transfer of the material to be treated. Electrical heaters immersed in the bed provide supplemental heat as required. The bed temperature is controlled closely to ensure volatilization and partial steam reforming of organic compounds. It also is controlled to ensure retention of radionuclides, including cesium compounds in solid form, along with other inorganics in the bed. The first stage is continuously fed waste material. Glass frit or sand is added as the startup bed material. With the exception of large inorganic chunks that may be in the feed material (these will be removed or not excavated), the bed material will be less than 0.05 m (2 in.) mean size material. Thus, size reduction of the feed material will be required. Any material fed to the steam reformer must also be capable of being entrained within a fluidized bed. As such, dense materials are not amenable to this treatment process.

e. References herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the U.S. Government, any agency thereof, or any company affiliated with the Idaho National Engineering and Environmental Laboratory.

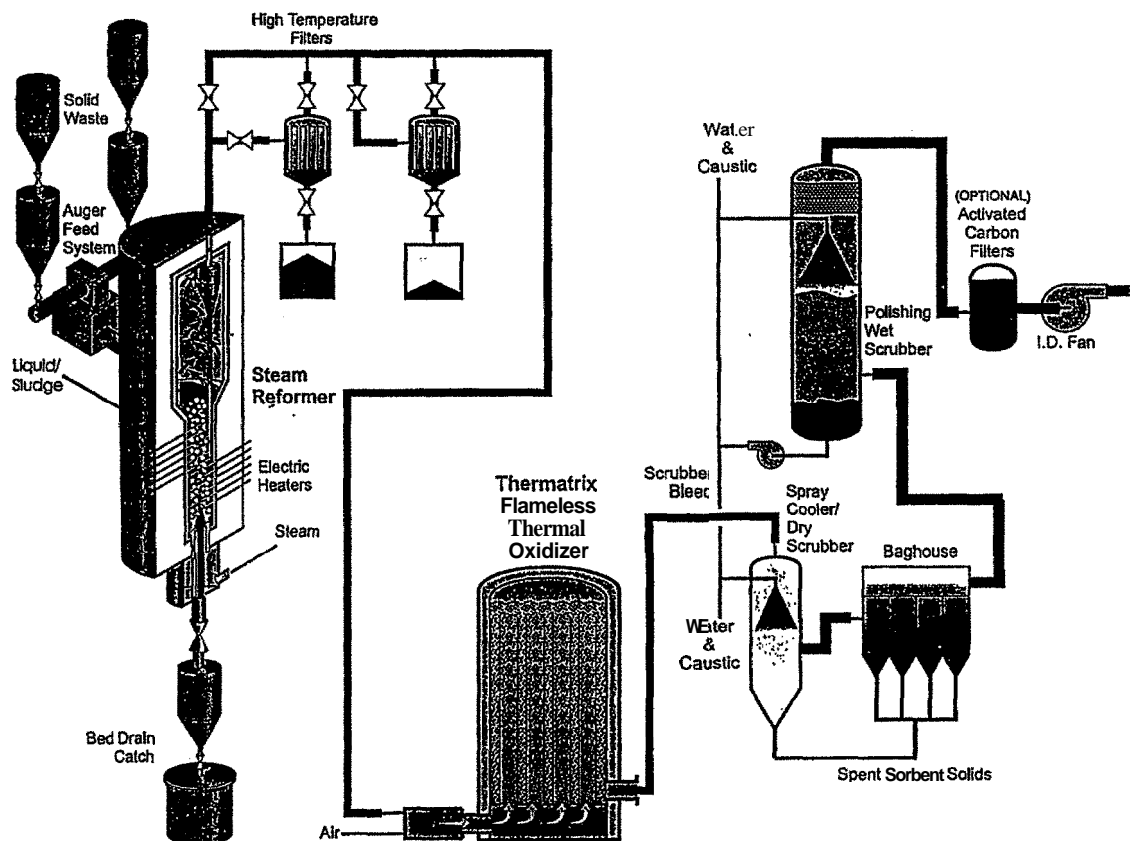


Figure 1. ThermoChem fluidized bed steam reformer.

An internal cyclone (caused by tangentially injected steam) and a high-temperature pulse jet high-efficiency particulate air (HEPA) filter at the exit of the first stage steam reformer eliminate the carryover of particulates in the synthesis gas stream. After particulate removal, the synthesis gas flows into a flameless thermal oxidizer. This nonincineration system converts carbon monoxide, hydrogen, and hydrogen vapors into carbon dioxide and acid gases. A second stage, higher temperature steam reformer (not shown in the figure) is available upstream of the thermal oxidizer and can be used if necessary to ensure higher destruction and removal efficiencies. A gas cleanup system removes acid gases as dry salts, and then an activated carbon filter polishes the affluent before discharge.

Testing on a 41-kg/hour (90-lb/hour) process development unit has verified complete retention of cesium and heavy metal compounds as a solid in the first stage reactor bed materials with volume reductions typically of 10 to one. Successful testing on low-level mixed waste surrogates have been performed on liquid waste streams. Destruction and removal efficiencies up to 99.9999% have been achieved for RCRA organic components found in U.S. Department of Energy waste.

The single-stage THOR pyrolysis and reforming process (used for volume and mass reduction of depleted ion exchange resins) has been reconfigured into a two-stage process to handle drummed waste. Figure 2 gives a flow diagram. The first stage comprises a sealed, electrically heated pyrolysis chamber (pyrolyzer) that encloses and heats the drums. The water content of the waste is evaporated, and organics are volatilized. A low-volume carbon residue is formed from most organic polymers, cellulose, and wood products. The inorganic components of the waste and solid pyrolyzed waste residues are retained in the drum. Pyrolysis gases pass from the drum pyrolyzer and are filtered to remove any trace particulates entrained in the pyrolyzer gas flow. Particulate carryover is very low as the pyrolysis gas volumes are

small. Because many particles are of submicron size, no steam is introduced into the drums, ensuring minimal carryover of particulates.

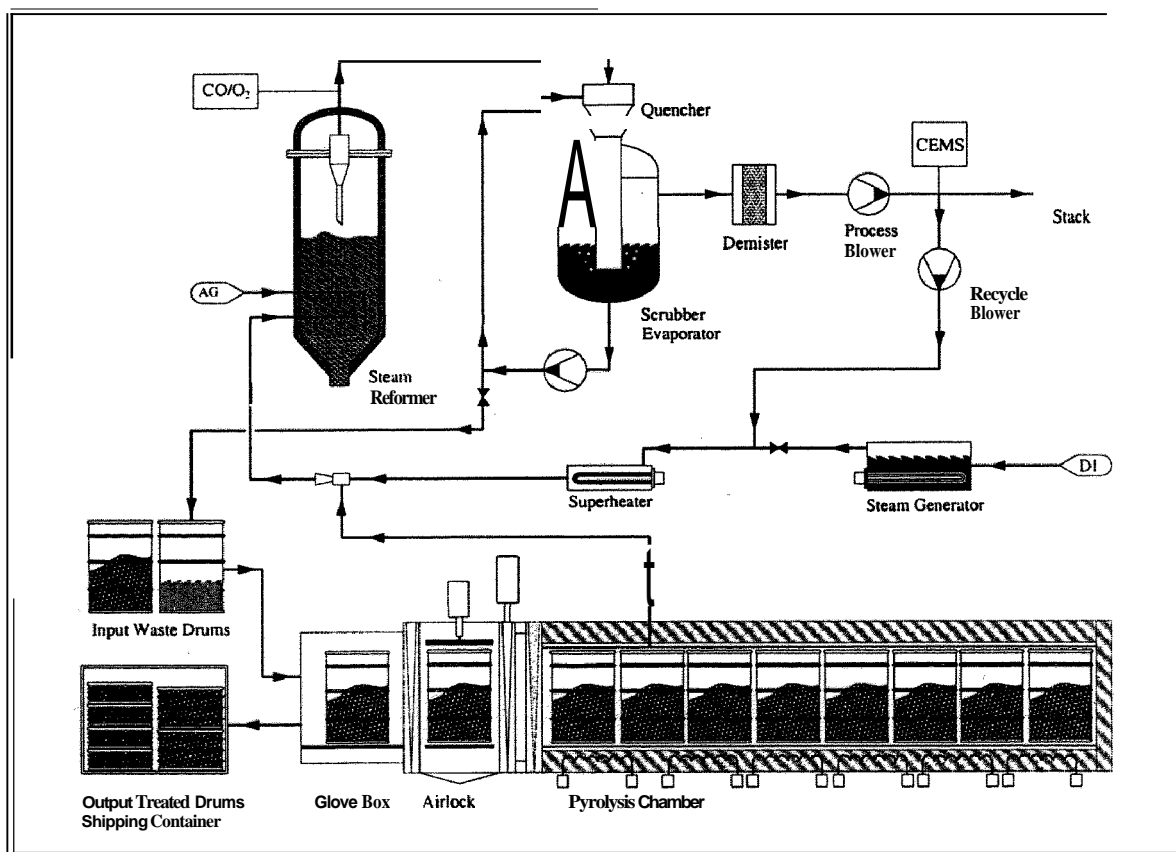


Figure 2. Thermal Organic Reduction pyrolysis steam-reforming system.

The gases from the pyrolyzer chamber are distributed into the bottom of the fluid bed steam reformer. Air is also injected (see AG in Figure 2) into the fluidized bed where volatile organics are converted into carbon dioxide and water vapor. Steam, superheated from 300 to 600°C, is used as the reforming medium. The fluidized steam, water vapor, and carbon dioxide gases exiting the reformer are then cooled in a water-cooled, thin-film quencher. The cooled off-gases are then scrubbed to remove and neutralize acid gases volatilized from waste in the drums.

The Duratek steam-reforming process can be used on drummed waste or loose, excavated material as illustrated in Figures 3a and 3b. The excavated waste is placed into a prereduction hopper that directs the waste to a slow-speed, high-torque shredder system that reduces waste to less than 2.5 cm (1 in.) in diameter (see Figure 3a). Then the feed material is transferred to a heated screw. The heated screw evaporator (HSE) continuously agitates the shredded waste material as it progresses through the screw housing. Electrical heating at the HSE inlet establishes a temperature of 315°C or higher. This temperature is increased through four distinct zones until 593°C is attained near the HSE outlet. By this time, the waste material has been volatilized completely.

Recirculated syngas from the HSE exhaust is introduced in the HSE housing to flow countercurrent to the waste material. This syngas is primarily CO, H₂, H₂O, volatile organics, and hydrocarbons generated

in the HSE by the high-temperature volatilization process. The syngas is pulled through the HSE by an induction fan that also maintains a negative pressure on the HSE.

The processed granular solid residue exits the HSE through a delumper that reduces any large aggregations to a sandy consistency. This residue is held in an exit hopper where it is discharged every few minutes to a residue collection drum for cooling. The temperature of the syngas exiting the HSE is maintained between 534 and 1,093°C. The steam-reformer reactions initiated in the HSE proceed rapidly to completion in the second-stage reformer (see Figure 3b) where elevated temperatures destroy the volatilized organics present in the syngas. In addition to accelerated reaction rates achieved in the steam reformer, the high temperatures provide the thermodynamic conditions necessary for high destruction and removal efficiencies.

The Duratek process also could be used in conjunction with a rotary kiln. In this scenario, solid waste is fed to a rotary kiln, hydrocarbons are pyrolyzed in the kiln, and the hydrocarbons are fed to the steam-reforming system. Duratek has formed a partnership with Technip of Germany to develop this integrated process.

4.1.2 Advantages, Disadvantages, and Issues

The advantages and disadvantages highlighted in the earlier evaluation (INEEL 1997) are still valid for steam reforming in general. One issue brought forward in the report stated that the Synthetica (now Duratek) process, by introducing steam at 300 to 600°C, acts more as an incinerator than as a steam-reforming process. High CO₂ and H₂O emissions from processing a 100% organic waste stream indicate this. A study of the thermodynamic equilibrium diagram for steam-reforming reactions shows that this is true. Duratek has since upgraded the system to restrict the access of O₂ (air infiltration) to allow true steam reforming in the heated screw evaporator.

4.1.3 Evaluation

4.1.3.1 Performance.

4.1.3.1.1 Range of Feed Material — Liquid and fines treatment can be accomplished by any of the steam-reforming technologies identified. However, this media is not the matrix of concern for the majority of OU 7-10 waste. Both Duratek and THOR Treatment Technologies can steam reform excavated waste that has been placed in 55-gal drums. Duratek also can steam reform excavated waste by passing it through its feed shredder and prefeed hopper.

4.1.3.1.2 Treatment Effectiveness — The treatment effectiveness for VOCs is given in the earlier evaluation (INEEL 1997). The VOCs identified in Table 2 should all be amenable to this process. The Duratek process has been shown to be very successful for thermal decomposition of sodium nitrate from the underground storage tanks at Hanford (Galloway, Dosch, and Sprung 1993). Conversations with Dr. Galloway of Duratek revealed that the steam-reforming process destroyed potassium ferrocyanide successfully and that it was easier to destroy than the nitrates.

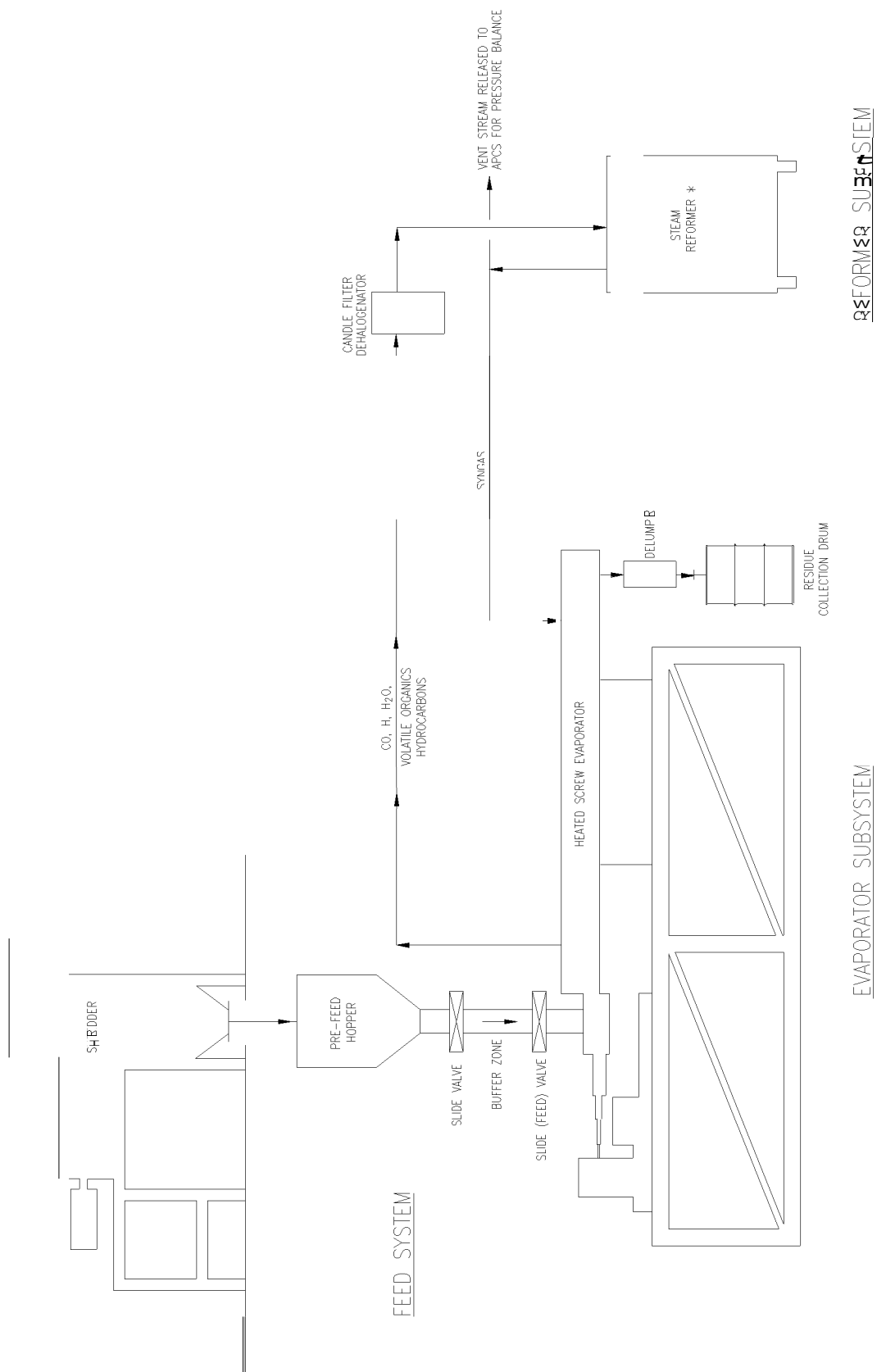
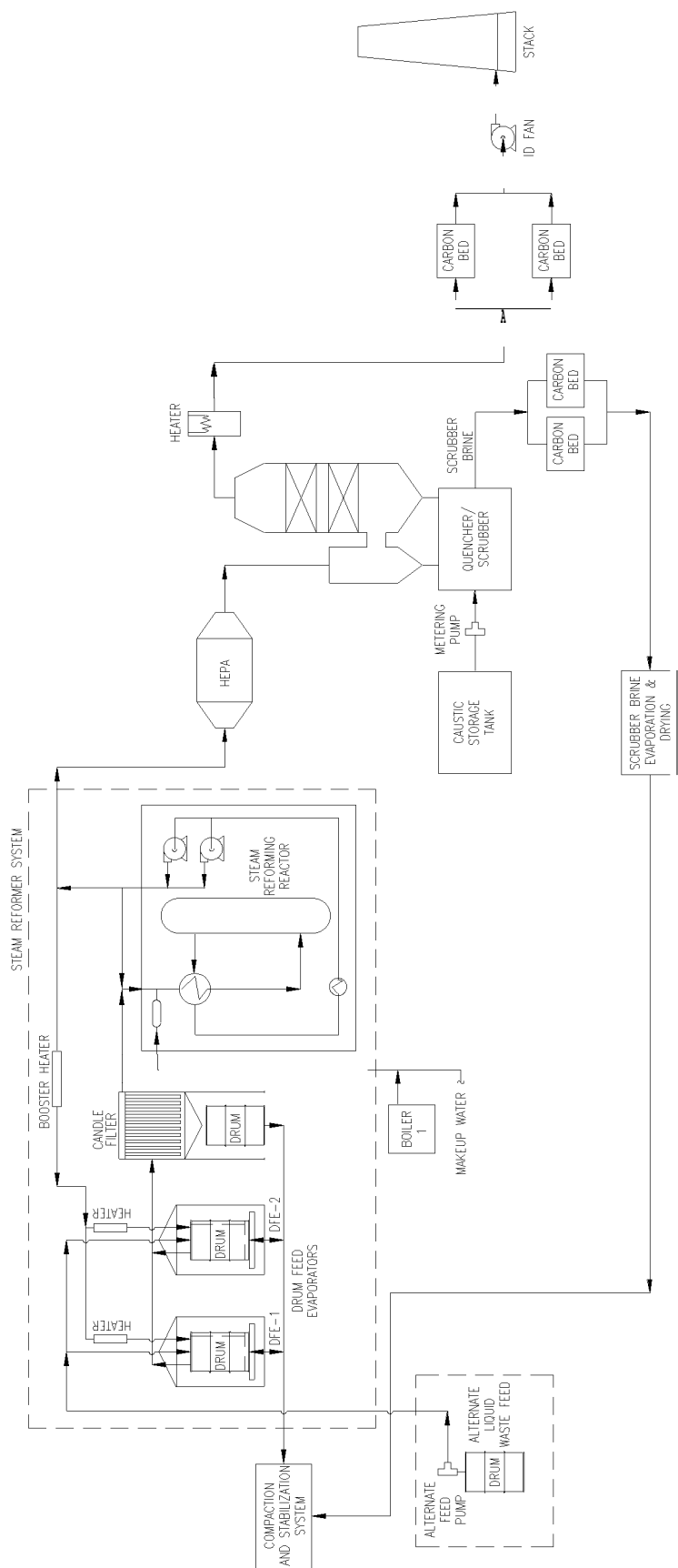


Figure 3a. Duratek steam-reforming system.



BEAR CREEK STEAM REFORMING
PROCESS FLOW DIAGRAM

Figure 3b. Duratek steam-reforming system (continued).

4.7.3.2 Readiness for Deployment.

4.1.3.2.1 Level of Development and Understanding — Studsvik (THOR Treatment Technologies) has a facility in Erwin, Tennessee, for processing radioactive resin beads from ion exchange systems. ThermoChem has conducted preliminary conceptual design for both 136- and 227-kg/hour (300- and 500-lb/hour) waste-processing units in addition to conducting extensive studies on a 4 l-kg/hour (90-lb/hour) unit for liquids and fines. Duratek has developed the Synthetica steam-reforming technology for the treatment of drummed and loose waste forms. In all cases, if steam-reforming technology is chosen as the preferred technology for treating the OU 7-10 waste, a processing facility will be constructed, and a permit will need to be obtained.

4.1.3.2.2 System Complexity — The earlier evaluation (INEEL 1997) fully identifies system complexities for each different type of steam-reforming system.

4.7.3.3 Environmental Health and Safety.

4.1.3.3.1 Emissions — The earlier evaluation (INEEL 1997) identifies emissions produced from steam-reforming systems.

4.1.3.3.2 Worker Health and Safety — The earlier evaluation (INEEL 1997) identifies health and safety issues associated with steam-reforming systems.

4.2 Thermal Desorption

Thermal desorption was not considered in the earlier evaluation (INEEL 1997) because the desorbed vapors were considered to be combusted in an afterburner, thus classifying the thermal desorbers as incinerators. However, thermal desorbers can be operated so that desorbed vapors can be condensed to recycle the volatilized material, or they can be destroyed through various methods (e.g., catalytic oxidation or flameless oxidation). Thermal desorption units can be operated in various configurations. They range from direct-fired hydrocarbon fuel systems to units heated by thermal screws operated by using hot oil.

4.2.1 Technology Description

One system with potential for treating the majority of OU 7-10 waste streams is the SepraDyne-Raduce high-vacuum rotary retort. This process, a simple separation technology, removes volatile compounds from nonvolatile matrices. SepraDyne-Raduce has developed, patented, and commercialized an indirectly heated rotary retort that operates on high vacuum and high temperature. The combination of these features produces an environment capable of volatilizing (1) water, (2) most organic compounds, and (3) low to moderate boiling point metals, such as mercury, arsenic, selenium, and cadmium, with near zero toxic air emissions. Depending on the waste matrix, it can reduce the waste matrix volume during treatment. Because air for combustion and gases to transport the products of combustion (sweep gases) are eliminated from the retort, combustion will not occur, and total gas volume exhausted to the atmosphere is minimized. Only volatilized material and gases initially present will exit the retort, which substantially reduces size of the off-gas treatment equipment required.

Figure 4 shows the operating parameters and processing sequence of the rotary vacuum retort. Mixed waste such as soil, sludge, and 55-gal drums are size reduced before being fed to the retort. Once loaded, the retort is sealed, and a vacuum of 0.6 m (25 in.) or more of Hg is established. The retort is set into rotation, and burners are ignited to heat the retort's exterior. Heat is applied indirectly within an insulated firebox by an arrangement of burners fueled by natural gas, diesel oil, or propane. As the

temperature of the mixed waste in the retort gradually increases, some substances will volatilize. For instance, volatile organics, water, sulfur, and then mercury will be transported sequentially out of the retort to the treatment and recovery system. The vapors diffuse out of the retort and are condensed in a cold-water impinger system. Because of the high-vacuum environment, no sweep gases are needed. Thus, virtually all volatilized substances are condensed to liquid in the impinger. The resultant impinger water can be treated to destroy condensed hydrocarbons by a wastewater treatment system such as electron beam or UV photo-oxidation. Mercury can be removed from the settling chamber and then amalgamated before disposal.

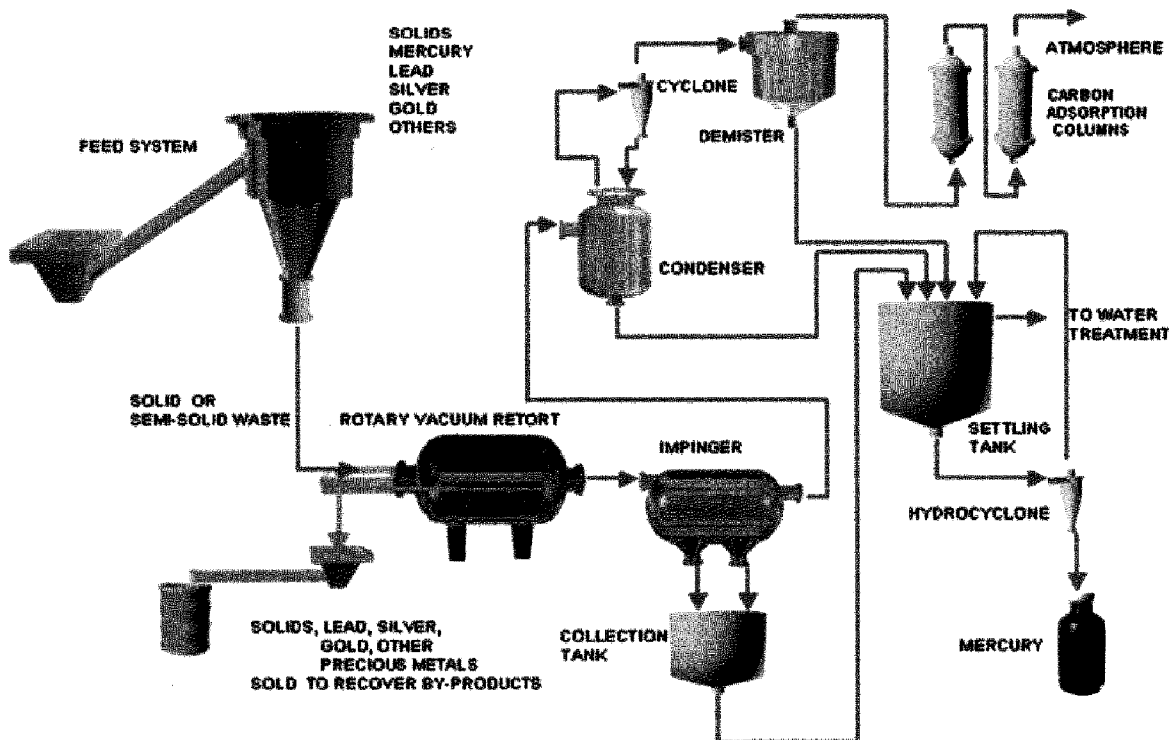


Figure 4. SepraDyne-Raduce high-vacuum thermal desorber.

The retort temperature is held at a moderate level to allow water to be removed from the matrix. When the drying phase is complete, operators raise the retort temperature to a target value, typically in the range of 600–740°C, and hold it there for a predetermined period. During this period, vacuum is kept at 0.5 m (20 in.) of Hg or higher. At this processing condition, any remaining organic compounds, including heavy tars, and all compounds of mercury are volatilized.

Chemicals are separated from the condensed water through a wastewater treatment train, and the water is treated for the contaminants present. The activated carbon columns and subsequent HEPA filters remove any trace hazardous vapors that have passed through the impinger system. The material is maintained in the retort for a predetermined process time at the target temperature until all COCs have been removed or pyrolyzed. After the process sequence, burners are turned off and the vacuum is released. The process material is then unloaded into a receiving vessel for stabilization (if RCRA inorganic compounds are above LDR values) and disposed back into OU 7-10.

Before processing an individual waste stream, treatability testing is performed to determine the operating condition (e.g., vacuum, hold temperature, or processing time) for that particular waste stream. Treatability testing can be performed on a small bench-scale unit that mimics the full-scale unit.

4.2.2 Advantages, Disadvantages, and Issues

A 1,814-kg- (2-ton-) per-batch unit currently is operating at a mine in Arizona recovering mercury from wet sludge filtrate from ore smelting. A two-drum-per-batch unit currently is being constructed for mixed waste operations. This unit will separate RCRA organics and volatile metals from mixed waste for further stabilization and disposal. Resource Conservation and Recovery Act organics can be thermally decomposed in the high-vacuum process, recovered from the process and recycled, or recovered from the process and destroyed in a catalytic or thermal oxidizer. A 91-kg- (200-lb-) per-batch unit has undergone extensive testing at Brookhaven National Laboratory.

The advantage of using a high vacuum, coupled with a relatively high-temperature thermal desorber, is that both volatile and semivolatile compounds can be removed from the contaminated matrix. The target contaminant groups for low-temperature thermal desorbers are nonhalogenated VOCs and fuels but not the COCs at OU 7-10. The proprietary seal that enables large vacuums to be formed is claimed to ensure that volatile radionuclides (Cs-137) would not be released but captured in the condenser system.

4.2.3 Evaluation

4.2.3.1 Performance.

4.2.3.1.1 Range of Feed Material—Ranking—Medium: This process is typical of many thermal treatment systems in that reactive or explosive material cannot be fed to the thermal desorber. Solid or semisolid material can be fed to the retort through the conventional feed system. This requires that no solids greater than 0.05 m (2 in.) in diameter can be fed to the system. Therefore, sizing of the waste feed may be required. A new two-drum-per-batch unit currently is being constructed for application to mixed alpha low-level and TRU waste. In this arrangement, two retorts are fed from a common feed system (shredder and auger) and discharge into a shared off-gas system. Figure 5 gives a schematic of the dual retort application to mixed waste. The new system is being designed for materials handling and contamination control. However, the constructed unit will have to be fully tested (including the seals) to ensure its operability in the OU 7-10 environment.

The sodium and potassium nitrates found in the RFP sludge are not anticipated to be a challenge for the desorber. Conversations with SeptraDyne personnel have indicated that these nitrates will be thermally destroyed to form oxides of nitrogen. Both the sodium and potassium cyanide, if encountered, are anticipated to thermally decompose in the desorber.

4.2.3.1.2 Treatment Effectiveness—Ranking—High: The Transuranic and Mixed Waste Focus Area conducted a series of demonstrations of the SeptraDyne-Raduce technology on various mercury and organically contaminated mixed waste (TMFA 2001). Testing was conducted on a 91-kg- (200-lb-) per-batch unit used for treatability testing. The test feed consisted of 1,383 kg (3,050 lb) of soils and animal carcasses that were processed during the demonstration. All processed material and soils were reduced to a fine homogeneous powder matrix with a final product weight of approximately 1,071 kg (2,360 lb). The data show that the vacuum thermal desorption process was able to provide a final product with a 23% weight reduction. Based on visual inspection, the volume reduction was estimated at approximately 40–50%. Additional testing on soil, animal carcasses, and dry active waste produced weight reductions up to 60%.

4.2.3.2.3 Emissions — Ranking — High: Emissions from the stack of the high-vacuum thermal desorption unit are minimized. This is because no sweep gas is used to move the organic and volatile inorganic compounds through the off-gas treatment train. Therefore, the volume of products to the off-gas treatment train is greatly reduced compared to other thermal systems. Any organic contamination in the impinger collection tank can be treated to acceptable standards using electron beam or UV photo-oxidation.

4.2.3.2.4 Worker Health and Safety — Ranking — Medium: Only minor adverse effects on worker health and safety are expected. The system operates at elevated temperatures and high vacuum. Suitable precautions must be taken while operating in these conditions.

5. NONTHERMAL TREATMENT TECHNOLOGIES

The earlier evaluation (INEEL 1997) has identified numerous nonthermal technologies for the treatment of OU 7-10 waste. However, one treatment process, which was neglected in that document and has potential for treating a wide variety of OU 7-10 waste, is dehalogenation of the organic-contaminated media. This technology has been identified by the Blue Ribbon Panel of experts in 2000 and is included as an extension of the earlier evaluation (INEEL 1997).

5.1 Chemical Dehalogenation

5.1.1 Technology Description

Commodore Solution Technologies, Inc. (Commodore) has developed a solvated electron dehalogenation process that removes halogens from organic-contaminated media and converts them to a nonhazardous high-molecular-weight hydrocarbon and a salt. Commodore has patented the Solvated Electron Technology (SET) to use in the SoLV process to dehalogenate organic compounds. Dissolving sodium metal in liquid anhydrous ammonia to form a solution, the solvated electron solution, activates the process. Similar solutions also can be formed by dissolving alkali or alkaline-earth metals, including sodium, calcium, lithium, and potassium, in anhydrous liquid ammonia. Formation of the solvated electron is believed to occur as illustrated in the following equation:



The solutions, which form rapidly when metal enters the ammonia, are characterized by a deep blue coloration and an electrical conductivity approaching that of liquid metals. This solvated electron solution can split the halogens from organic contamination, yielding measurable amounts of the halogen anion. In the case of aromatic material, the parent hydrocarbon can react further to produce high-molecular-weight compounds. In addition to halogens, many other organic molecules are reactive towards solvated electrons. Organic phosphorous and sulfur compounds, such as pesticides and chemical warfare agents, are known to be reactive to solvated electrons.

In addition to destroying organic contaminants, the SoLV process can remove metals that may contaminate soil and other matrices. One of several patented postprocesses accomplishes this. One process is removing inorganic compounds by washing the treated matrix with a mixture of ammonia and water at high pH using in-situ-generated sodium hydroxide.

For normal operations processing soil, or other small-grained material, waste is fed to the reactor, followed by sodium and liquid ammonia (see Figure 6). When processing larger, solid materials (up to 0.56 m [22 in.] in diameter) in the reactor, the solid material and liquid ammonia are mixed. The ammonia washes the contamination from the substrate. After a brief period of additional mixing, sodium metal is added to the ammonia slurry by solid or molten addition. Electrons are freed from the sodium and chemically destroy the contaminants. When the process is complete, ammonia is retained for reuse, and treated material is released to the environment. Wet sludge requires a water removal step before using the process to destroy the contaminant. Drying or using a prewash module accomplishes water removal.

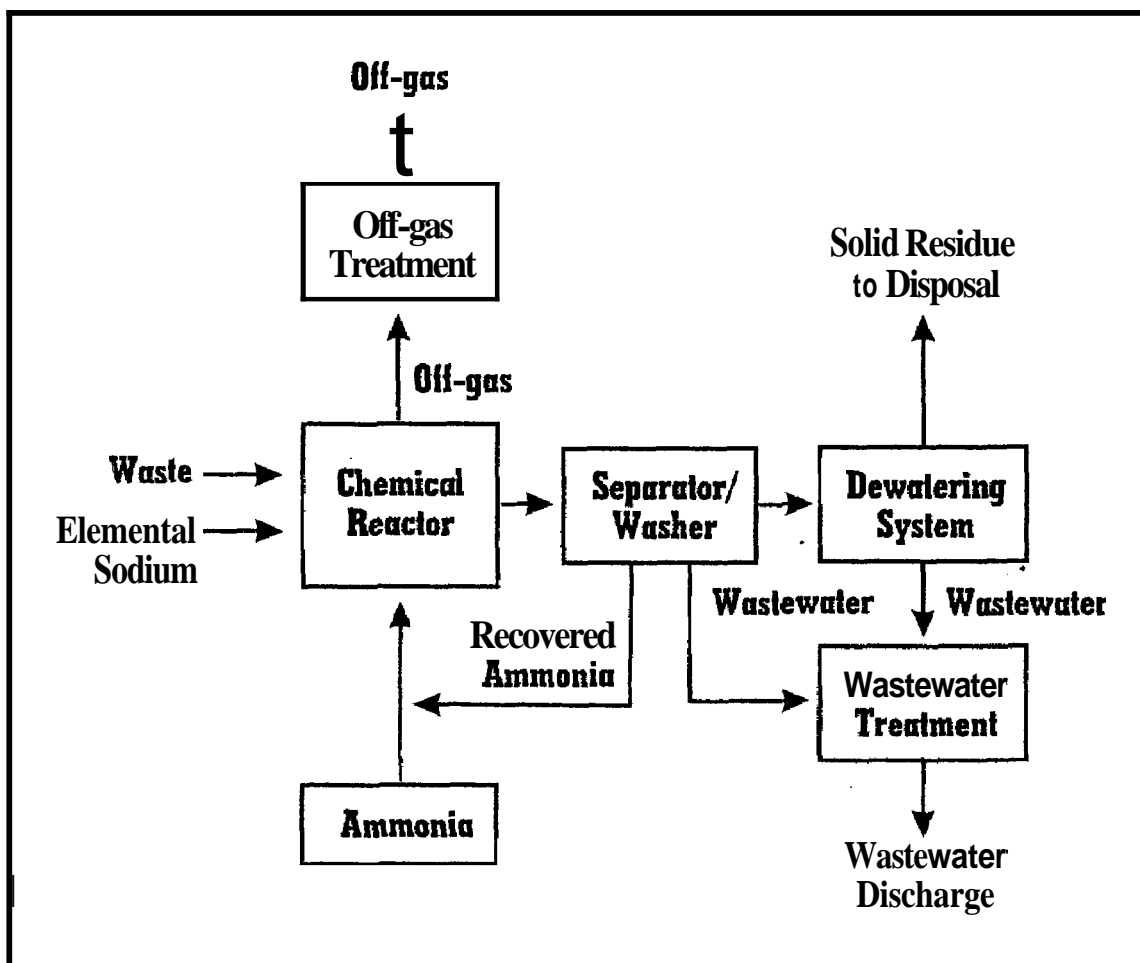


Figure 6. Commodore Solvated Electron Technology treatment process.

5.1.2 Advantages, Disadvantages, and Issues

Advantages

- The process is a well-established, commercial-scale technology
- The technology has been shown to be effective in the destruction of PCBs, dioxins/furans, halogenated compounds, chlorofluorocarbons, polyaromatic hydrocarbons, pesticides, explosives, and mixed waste
- The process has partial success on removing metals from contaminated soils
- Reactions are conducted at below 4°C, thus protecting against volatile emissions
- The destruction process is carried out in a totally closed system
- End products from the process are metals, salts (sodium chloride), and long-chain hydrocarbons (not RCRA regulated) that are released to the environment.

Disadvantages

- Moisture levels greater than approximately 40% can cause processing problems. To ensure that no wastewater is produced from the process, a drying module developed by Commodore can be installed on the feed system. This module separates the water from the waste stream and directs any separated organics back into the treatment cell.
- The presence of long-chain hydrocarbons may cause complications in stabilizing waste streams with high levels of RCRA inorganic compounds. Another processing step may be required to destroy or separate the long-chain hydrocarbons from the final waste streams.
- The presence of RCRA organic compounds in excess of 20,000 ppm requires that the contaminated matrix be passed through the SET process at least twice. This increases the cost of remediation.

5.1.3 Evaluation

5.1.3.1 Performance.

5.1.3.1.1 Range of Feed Material—Ranking—High: The process is well known to treat various RCRA hydrocarbons in the feed. Highly chlorinated RCRA organics, such as PCBs and carbon tetrachloride, are readily treated using this process. Treating the RCRA inorganic component of the waste stream also has been successful. Metal nitrates are also amenable to treatment using this technology as they are converted to nitrites. This treatment process is not anticipated to have any effect on the cyanides that may be in the waste stream. The moisture content of the waste stream must not exceed 40%.

5.1.3.1.2 Treatment Effectiveness—Ranking—High–Medium: The conversion of RCRA-listed hydrocarbons to long-chain benign hydrocarbons is shown to be very effective. However, the presence of long-chain hydrocarbons may cause stabilization problems with RCRA inorganic-contaminated waste streams.

This technology does not produce a volume reduction. The process does not destroy the RCRA organic components or reduce the mass and volume of the waste matrix; it converts the RCRA organic components to benign products.

5.7.3.2 Readiness for Deployment.

5.1.3.2.1 Level of Development and Understanding—Ranking—High: The Commodore SoLV is a mature technology with approximately 20,000 hours of operating experience in 14 states on multiple tons of material. The Environmental Protection Agency has issued a nationwide Toxic Substances Control Act treatment permit for SET for mobile PCB chemical destruction in soils, metals, oils, organics, and debris. The process recently completed remediation of 400 tons of PCB-contaminated soil in Pennsylvania at a feed rate of 1,814 kg (4,000 lb) per batch. The technology has been used on mixed waste at Weldon Spring, Missouri, where PCBs, chlorinated, and aromatic compounds were greater than 97% destroyed. In fact, the chlorinated compounds were greater than 99.99% destroyed. Treatability testing at Brookhaven National Laboratory reduced levels of Freon 113; 1,1,1 trichloroethane; and trichloroethene from hundreds of ppm levels to nondetectable levels. More recently, thirty-one 55-gal drums of Freon still bottoms from Oak Ridge National Lab contaminated with Freon 113; 1,4 dioxane; and other contaminants were treated to meet land disposal requirements. One drum treated contained over 50% mass of target compounds.

Treatability testing of the Commodore system was conducted at the Envirocare of Utah facility in Clive, Utah. The system removed the COCs in a sludge mixed waste. The contaminants in the sludge were Freon 113; 1,1,1 trichloroethane; and trichloroethylene with a moisture content of 75%. After processing in such a wet waste stream, the final level of these three analytes was below the universal treatment standard for the particular analyte. In addition, the toxicity characteristic leaching procedure value for lead was reduced by two orders of magnitude to regulatory requirements. The chlorinated compounds were broken down into carboxylic acids. If stabilization of the sludge was required for stabilization of inorganic compounds, these carboxylic acids would not hinder the stabilization process. However, treatability testing of the treated matrix would be required.

Commodore's SoLV technology was evaluated in the U.S. Environmental Protection Agency Superfund Innovative Technology Evaluation Program. During the demonstration, the SoLV process treated 30 batches of PCB-contaminated soil. Of these 30, 21 treated batches of soil showed greater than 99% removal of PCBs. Only one batch of the remaining nine demonstrated less than 90% PCB removal. The demonstration was not able to evaluate a caustic wash process to remove metals because Commodore was not able to make the necessary equipment modifications to recover water from the caustic washes. The process was successful in reducing polyaromatic hydrocarbons to below detection limits. The technology's effectiveness increased as the soil particle size decreased. This means the process is more effective on silt and clays than on coarser particles.

A mobile 1,814-kg- (4,000-lb-) per-batch system could again be located either next to OU 7-10 or next to a size reduction facility. The mobile nature of the Commodore system should make facility location easy.

5.1.3.2.2 System Complexity—Ranking—Medium: The system complexity is not as great as many thermal processes, but because of ammonia and sodium required for the reaction, only qualified personnel should operate the equipment. The chemical reactions are completed in a short time; recovery of the ammonia and waste loading and discharge requires the most time. Qualified personnel should perform these operations.

5.7.3.3 Environmental Health and Safety.

5.1.3.3.1 Emissions—Ranking—High: The SET process operates as a closed loop system except when occasionally venting small quantities of hydrogen. Hydrogen enters the system when adding waste to the treatment cell through the inlet port. This release of hydrogen also leads to the release of trace amounts of ammonia. The human nose can detect very low concentrations of ammonia, but the levels are not high enough to be of concern.

Because of the low temperature of the dehalogenation process, little off-gas is generated. The low system temperature also means radioactive particles are not likely to be carried up into the off-gas.

5.7.3.3.2 Worker Health and Safety—Ranking—High: A misconception with solvated electron dehalogenation is the safety concern about potentially explosive conditions in the treatment cell and with the handling of corrosive process solutions. The Commodore SET process has been in operation for over 15,000 hours without any incident involving explosions or the handling of corrosive materials. Commodore received a safety award at Weldon Spring after treating 45,359 kg (100,000 lb) of mixed waste.

6. COMPREHENSIVE ENVIRONMENTAL RESPONSE, COMPENSATION, AND LIABILITY ACT SCREENING CRITERIA

The Comprehensive Environmental Response, Compensation, and Liability Act of 1980 has resulted in a guidance document for selecting technologies that should be considered for treatment of Comprehensive Environmental Response, Compensation, and Liability Act sites. This document, *Guidance for Conducting Remedial Investigations and Feasibility Studies Under CERCLA*, (EPA 1988) suggests that potential technologies be screened based on effectiveness, implementability, and cost. The technologies identified in this report are screened using these criteria.

6.1 Effectiveness

Steam reforming received a high ranking in the selection process described in the 1997 evaluation (INEEL 1997). The process is anticipated to be effective in destroying volatile and semivolatile RCRA organics found in the OU 7-10 waste streams. Treatability testing will be required to determine that no RCRA-regulated products are formed in the syngas or from destruction of the syngas. For the ThermoChem process, it will be necessary to determine the frequency of bed regeneration or replacement and secondary waste generation associated with contaminated bed material. It may be necessary to determine if a fluidized bed process is a suitable treatment for the range of OU 7-10 contaminated matrices. Disposition of secondary waste generated during processing (i.e., spent carbon and scrubber blowdown) will have to be considered for all three steam-reforming processes.

The SeptraDyne-Raduce thermal desorber should be effective in separating organic contamination from the waste matrix. The parameters of operation (time, vacuum, and temperature) should determine effectiveness of the desorption process and volume reduction of the host matrix. If RCRA organics are desorbed and collected from the desorber, a disposition path for the reclaimed material needs to be determined. This may include a thermal oxidizer to treat the reclaimed organics. Disposal of waste generated during the process will have to be taken into account.

The Commodore SET process should be effective in reducing halogenated and other RCRA organics into less complex, non-RCRA, long-chain hydrocarbons. However, this may provide problems when stabilizing the treated product for RCRA-listed inorganic compounds. Treatability testing on each matrix and type of contaminant is required.

An electron beam or UV photo-oxidation process could be coupled with each of these processes to destroy organic contaminants in the wastewater generated. Treatability testing has shown that carbon tetrachloride can be reduced in liquid waste streams by greater than 99% using these processes.

6.2 Implementability

The ThermoChem steam-reforming unit primarily is intended for the reforming of liquid or other matrices that can be supported in a fluidized bed. This only accounts for less than 7% of the waste requiring treatment at OU 7-10. The THOR process can be used for excavated, drummed waste. The Duratek system can be used for either drum waste or loose debris. A shredder placed above the feed hopper sizes the feed material.

If a steam-reforming process is chosen as (or one of) the preferred alternative(s), a processing system would have to be constructed at a location next to OU 7-10. Full-scale systems exist at fixed locations; portable units have not yet been constructed. Scheduling and budgeting would have to be considered in constructing and permitting a treatment process using steam reforming.

The SepraDyne-Raduce system is a trailer-mounted unit that can be erected at any suitable location. Power and potable water are required for operation. Both are readily available at the Subsurface Disposal Area. Size reduction is required for any feed material greater than 2 in. in diameter fed through the existing hopper feed system. A mixed waste treatment system currently is being designed to thermally desorb two 55-gal drums per batch. Once constructed, this unit can be implemented easily at OU 7-10.

The Commodore SET system is available in various sizes, depending on the throughput of waste treatment required. Sizing of the waste material may be required to fit into the treatment reactor. Alternatively, large debris can be washed with ammonia, and the ammonia (with contaminants) can be transferred to the reactor for treatment. All units are trailer mounted, needing a relatively small footprint. The company does possess a nationwide Toxic Substances Control Act permit for SET for PCB chemical destruction in soils, metals, oils, organics, and debris.

6.3 Cost

The figures presented in this document are estimates to show the relative economic feasibility of each treatment process. A detailed economic analysis would have to be performed by each of the technology vendors to provide a defensible cost estimate.

The cost estimate for steam reforming is considered the same order of magnitude as that for incineration. This was confirmed during discussions with Duratek personnel. An economic analysis was performed on the ThermoChem steam-reforming technology. Results of this analysis gave an estimate of \$11/kg (\$5/lb) based on a 136-kg/hour (300-lb/hour) unit (ThermoChem 1998).

The capital cost for the main processing equipment of the SepraDyne-Raduce unit is considered to be 50% of a similar-sized incineration process. However, feed preparation and final waste form processing and dispositioning costs are considered equal because pre and postprocessing requirements of both systems are the same. Operating costs for the off-gas system are considerably lower for the high-vacuum thermal desorber because of the reduced gas flows. Hence, there is less particulate carryover. Carbon adsorbers, HEPA filters, and impinger solutions in the desorber have a greater operating lifetime for the same feed rate compared to incineration off-gas systems. If the desorber is operated in the pyrolyzing mode, the off-gas system must be capable of measuring carbon monoxide, particulates, hydrochloric acid, and nitrous oxide. This will increase the cost of operating the off-gas system.

Discussions with Commodore personnel have revealed that the SET process can be used to treat mixed waste for a cost of \$18.70/kg (\$8.50/lb). As stated earlier, RCRA organics concentrations greater than 20,000 ppm require additional passes through the system. Thus, if the waste batch requires an additional run through the system, the cost will be at least \$37.40/kg (\$17/lb).

7. CONCLUSIONS

Two thermal technologies and one chemical extraction technology have been identified for treating and removing RCRA organic contamination from OU 7-10 waste streams. Steam reforming was identified in *Evaluation of Alternative Nonflame Technologies for Destruction of Hazardous Organic Waste* (INEEL 1997) as a highly ranked technology for further investigation. Three steam-reforming technologies were investigated for their application to removing organic contamination in the OU 7-10 waste. The investigation revealed that at least two technologies (THOR and Duratek) are developed sufficiently for treatability testing on OU 7-10 waste surrogates, and waste forms can be initiated. The ThermoChem unit is a fluidized bed system that may have complications with dense objects (cemented sludge) even after size reduction to less than 0.05 m (2 in.) in diameter. The ability to maintain a fluidized bed with dense objects in the feed is questioned. The fate of cesium needs further investigation because of its volatility at 400°C. It is not expected that dioxin formation is likely to be a major concern because of the low presence of oxygen in the system. Emissions testing should be performed to investigate this.

A high-vacuum thermal desorption technology has been identified that could treat the large majority of OU 7-10 organic waste streams. Through its proprietary vacuum system, SepraDyne-Raduce has developed an indirectly fired desorption process that can separate organic and volatile inorganic contaminants from the host matrix and recondense them into a matrix that is easily treated. A two-55-gal-drum-per-batch unit presently is being constructed for application to mixed waste. Smaller units are also available for treatability testing.

The nonthermal technology investigated is the Commodore SET used in the SoLV process. This process chemically breaks down RCRA-listed halogenated hydrocarbons into common salts and long-chain, inert hydrocarbons. The process is commercially available in a variety of sizes ranging from small-scale treatability testing to full-scale remediation. The SoLV process also has been shown to reduce the level of inorganic contaminants in waste streams with the addition of generated sodium hydroxide. The ability of this process to treat a waste stream contaminated with both organic and inorganic compounds and to produce a waste stream amenable to stabilization needs to be demonstrated.

The three types of technologies highlighted in this report have all been used in a mixed low-level waste environment for use in either a full-scale remediation or at pilot-scale demonstration studies. However, using these technologies in a TRU environment such as OU 7-10 has yet to be demonstrated. Any technology or combination of technologies selected for the OU 7-10 remediation would have to allow for operation in full containment and be capable of processing a heterogeneous waste stream in terms of waste matrices, RCRA components, and radiological concerns.

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